Angular momentum of non-paraxial light beam: Dependence of orbital angular momentum on polarization

Chun-Fang Li*

Department of Physics, Shanghai University, 99 Shangda Road, Baoshan District, 200444 Shanghai, P. R. China

(Dated: September 12, 2009)

Abstract

It is shown that the momentum density of free electromagnetic field splits into two parts. One has no contribution to the net momentum due to the transversality condition. The other yields all the momentum. The angular momentum that is associated with the former part is spin, and the angular momentum that is associated with the latter part is orbital angular momentum. Expressions for the spin and orbital angular momentum are given in terms of the electric vector in reciprocal space. The spin and orbital angular momentum defined this way are used to investigate the angular momentum of nonparaxial beams that are described in a recently published paper [Phys. Rev. A 78, 063831 (2008)]. It is found that the orbital angular momentum depends, apart from an l-dependent term, on two global quantities, the polarization represented by a generalized Jones vector and a new characteristic represented by a unit vector $\mathbf{I}$, though the spin depends only on the polarization. The polarization dependence of orbital angular momentum through the impact of $\mathbf{I}$ is obtained and discussed. Some applications of the result obtained here are also made. The fact that the spin originates from the momentum density that has no contribution to the net momentum is used to show that there does not exist the paradox on the spin of circularly polarized plane wave. The polarization dependence of both spin and orbital angular momentum is shown to be the origin of conversion from the spin of a paraxial Laguerre-Gaussian beam into the orbital angular momentum of the focused beam through a high numerical aperture.

PACS numbers: 42.60.Jf, 42.25.-p

* Email address: cfli@shu.edu.cn

I. INTRODUCTION

The orbital angular momentum (AM) of light did not draw much attention [1, 2] until 1992 when Allen and his co-researchers [3] showed that a beam of Laguerre-Gaussian mode can carry both spin and orbital AM. They found that the spin is carried by the polarization σ and the orbital AM is carried by the helical wave front represented by a phase factor $\exp(il\phi)$, where l is an integer. Since then great progress has been made [4] in experiments. The orbital AM has been measured [5, 6]. The transfer of spin and orbital AM to microscopic particles [7, 8, 9, 10] and to liquid crystals [11, 12] has been observed.

Recently, experimentalists [13, 14] showed that the spin and orbital AM of a non-paraxial beam play distinct roles in the interaction with microscopic birefringent particles trapped off the beam axis in optical tweezers. It was observed [14] that the spin of light makes the particle rotate around its own axis and the orbital AM makes the particle rotate around the beam's axis. Furthermore, partial spin of a paraxial beam was observed [15] to be converted into orbital AM of a non-paraxial beam by a high numerical aperture. Those experimental results demonstrate that the spin and orbital AM of a non-paraxial beam are different in nature on one hand and are connected somehow to each other on the other. But up till now, there is no satisfactory theory to elucidate the difference and relation. The distinction that the spin is carried by the polarization and the orbital AM is carried by the helical wave front was drawn basically from the knowledge of a type of paraxial beams [3, 16, 17]. It is not valid for non-paraxial beams [18, 19, 20]. With a specific non-paraxial beam, Barnett and Allen [18] found that "the seemingly natural separation of the angular momentum...is no longer possible". The purpose of this paper is to advance a theory to explain the difference and relation between the spin and orbital AM of nonparaxial beams.

To this end, we should first know how to represent a nonparaxial beam that as a whole is in a definite state of polarization. As mentioned before, Barnett and Allen [18] once put forward a nonparaxial solution. But that solution was shown [21] to fail to meet the demand. Fortunately, a theoretical representation that meets the demand was recently developed [22]. The beam in this representation exhibits as a whole a definite polarization in the sense that all the plane waves that constitute the beam are described by the same normalized Jones vector. In other words, the normalized Jones vector in this representation is a global characteristic that plays the role of describing the polarization of the beam. This

Jones vector will be referred to as the generalized Jones vector. Apart from the global generalized Jones vector, a non-paraxial beam in this representation shows another global characteristic denoted by a unit vector. The global unit vector was applied [23] to explain the spin Hall effect of light [24]. In this paper, I will make use of this representation to show how the orbital AM depends on the polarization through the impact of the global unit vector.

Secondly, we should also know how to define the spin and orbital AM of an electromagnetic field in free space. The total AM $\mathbf{J}(\mathbf{x}_0)$ of a free electromagnetic field with respect to the point $\mathbf{x}_0$ is defined as [25]

$$\mathbf{J}(\mathbf{x}_0) = \int \mathbf{j} d^3x = \mathbf{J}(0) - \mathbf{x}_0 \times \int \mathbf{p} d^3x, \tag{1}$$

where $\mathbf{j} = (\mathbf{x} - \mathbf{x}_0) \times \mathbf{p}$ is the AM density with respect to the same reference point, $\mathbf{p} = \varepsilon_0 \mu_0 \mathcal{E} \times \mathcal{H}$ is the momentum density defined in terms of the electric vector $\mathcal{E}$ and the magnetic vector $\mathcal{H}$, and

$$\mathbf{J}(0) = \int \mathbf{x} \times \mathbf{p} d^3x \tag{2}$$

is the AM with respect to the origin. The separation of total AM into spin and orbital AM was discussed before [25, 26, 27, 28] by performing the integration in Eq. (1) by parts and neglecting a surface integral at infinity. In this paper, I will put forward a rigorous approach to the separation of total AM into spin and orbital parts by examining the property of momentum density. This approach allows us to apply the obtained result to plane waves.

The paper is arranged as follows. In Section II, it is found from the transversality condition that the momentum density of an electromagnetic field in free space splits into two parts. One part does not have any contribution to the net momentum; the other part produces all the momentum. The AM that originates from the former part does not depend on the choice of the reference point and is the spin. The AM that originates from the latter part is in general dependent on the choice of the reference point and is the orbital AM. In Section III, the integral expressions for the spin and orbital AM obtained in Section II are used to investigate the AM properties of nonparaxial beams described by the aforementioned representation. Since the light beam is assumed to be monochromatic, both the integrals of spin and orbital AM are infinite. In order to deal with the infinity, the technique of δ-function normalization is used. As expected, the spin AM is found to be dependent on the polarization. But what is surprising is that the orbital AM is also dependent on the

polarization. It is shown how the orbital AM depends on the polarization through the impact of the global unit vector. Two different problems are discussed in Section IV by making use of the obtained results. Section V concludes the paper with further remarks.

II. SEPARATION OF THE TOTAL AM INTO SPIN AND ORBITAL AM

Consider an arbitrary electromagnetic field in free space. Its electric vector in real space can be expressed as an integral over the plane-wave spectrum,

$$\mathcal{E}(\mathbf{x},t)=\frac{1}{2}\left\{\frac{1}{(2\pi)^{3/2}}\int \mathbf{E}(\mathbf{k})\exp[i(\mathbf{k}\cdot\mathbf{x}-\omega t)]d^3k+c.c.\right\}, \tag{3}$$

where $\mathbf{k}$ is the wave vector and $\mathbf{E}(\mathbf{k})$ is the electric vector in reciprocal space. The magnetic vector of the beam is derived from Eq. (3) and Maxwell's equations to be

$$\mathcal{H}(\mathbf{x},t)=\frac{1}{2}\left\{\frac{1}{(2\pi)^{3/2}}\int \frac{\mathbf{k}\times\mathbf{E}}{\mu_0\omega}\exp[i(\mathbf{k}\cdot\mathbf{x}-\omega t)]d^3k+c.c.\right\}. \tag{4}$$

Integral expression (3) or (4) leads to the following transformations [25],

$$\omega(-\mathbf{k})=-\omega(\mathbf{k}),\quad \mathbf{E}(-\mathbf{k})=\mathbf{E}^*(\mathbf{k}). \tag{5}$$

With the help of Eqs. (3) and (4) and vector algebra $\mathbf{a}\times(\mathbf{b}\times\mathbf{c})=(\mathbf{a}\cdot\mathbf{c})\mathbf{b}-(\mathbf{a}\cdot\mathbf{b})\mathbf{c}$, the momentum density splits into two parts,

$$\mathbf{p}=\varepsilon_0\mu_0\mathcal{E}\times\mathcal{H}=\mathbf{p}_1+\mathbf{p}_2, \tag{6}$$

where

$$\begin{aligned}\mathbf{p}_1 &= \frac{\varepsilon_0}{4(2\pi)^3}\int\frac{\mathbf{E}'\cdot\mathbf{E}}{\omega}\mathbf{k}e^{i(\mathbf{k}'+\mathbf{k})\cdot\mathbf{x}}e^{-i(\omega'+\omega)t}d^3k'd^3k\\ &\quad+\frac{\varepsilon_0}{4(2\pi)^3}\int\frac{\mathbf{E}'\cdot\mathbf{E}^*}{\omega}\mathbf{k}e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{x}}e^{-i(\omega'-\omega)t}d^3k'd^3k+c.c.,\end{aligned} \tag{7}$$

$$\begin{aligned}\mathbf{p}_2 &= -\frac{\varepsilon_0}{4(2\pi)^3}\int\frac{\mathbf{E}'\cdot\mathbf{k}}{\omega}\mathbf{E}e^{i(\mathbf{k}'+\mathbf{k})\cdot\mathbf{x}}e^{-i(\omega'+\omega)t}d^3k'd^3k\\ &\quad-\frac{\varepsilon_0}{4(2\pi)^3}\int\frac{\mathbf{E}'\cdot\mathbf{k}}{\omega}\mathbf{E}^*e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{x}}e^{-i(\omega'-\omega)t}d^3k'd^3k+c.c.,\end{aligned} \tag{8}$$

$\mathbf{E}\equiv\mathbf{E}(\mathbf{k})$, $\mathbf{E}'\equiv\mathbf{E}(\mathbf{k}')$, $\omega\equiv\omega(\mathbf{k})$, and $\omega'\equiv\omega(\mathbf{k}')$. Based on the transversality condition $\mathbf{k}\cdot\mathbf{E}=0$, it is readily proven by use of transformations (5) that $\mathbf{p}_2$ does not have any contribution to the net momentum,

$$\mathbf{P}_2=\int\mathbf{p}_2d^3x=0. \tag{9}$$

This tells us a fact that all the momentum $\mathbf{P}$ comes only from $\mathbf{p}_1$,

$$\mathbf{P} = \mathbf{P}_1 = \int \mathbf{p}_1 d^3x = \varepsilon_0 \int \frac{\mathbf{E}^* \cdot \mathbf{E}}{\omega} \mathbf{k} d^3k, \tag{10}$$

which is independent of time.

Accordingly, the total AM also splits into two parts,

$$\mathbf{J}(\mathbf{x}_0) = \int (\mathbf{x} - \mathbf{x}_0) \times \mathbf{p} d^3x = \mathbf{S}(\mathbf{x}_0) + \mathbf{L}(\mathbf{x}_0).$$

Because of property (9), the first part $\mathbf{S}$ that originates from momentum density $\mathbf{p}_2$ is independent of the choice of the reference point,

$$\mathbf{S}(\mathbf{x}_0) = \mathbf{S}(0) = \int \mathbf{x} \times \mathbf{p}_2 d^3x. \tag{11}$$

In other words, the fact that $\mathbf{S}$ is independent of the choice of the reference point roots in an intrinsic property of the electromagnetic field, the transversality condition. It is thus reasonable to regard this intrinsic AM as the spin. The second part that originates from momentum density $\mathbf{p}_1$ is in general dependent on the choice of the reference point,

$$\mathbf{L}(\mathbf{x}_0) = \mathbf{L}(0) - \mathbf{x}_0 \times \mathbf{P}_1, \tag{12}$$

where

$$\mathbf{L}(0) = \int \mathbf{x} \times \mathbf{p}_1 d^3x. \tag{13}$$

It is plausible to regard this part as the orbital AM. Substituting Eq. (8) into Eq. (11), one obtains by straightforward calculations

$$\mathbf{S} = \int \frac{\varepsilon_0}{i\omega} \mathbf{E}^* \times \mathbf{E} d^3k. \tag{14}$$

The momentum density $\mathbf{p}_2$ leads to the spin AM, though it does not produce any momentum. Such an astonishing fact means that there is no paradox on the spin AM of circularly polarized plane waves. This will be discussed in Section IV. Substituting Eq. (7) into Eq. (13), one has

$$\mathbf{L}(0) = \int \frac{\varepsilon_0}{i\omega} \mathbf{E}^\dagger (\mathbf{k} \times \nabla_\mathbf{k}) \mathbf{E} d^3k, \tag{15}$$

where $\nabla_\mathbf{k}$ is the gradient operator with respect to $\mathbf{k}$, and the superscript † stands for the conjugate transpose [29]. For the readers' convenience, the details to calculate Eqs. (14) and (15) are summarized in Appendix. It is very interesting to note that the spin (14) and

orbital AM (15) obtained this way look very like their quantum-mechanical counterparts [25].

At last, let us give here for later convenience the total energy of the beam in terms of the plane-wave spectrum,

$$W = \int (\frac{\varepsilon_0}{2}\mathcal{E}^\dagger\mathcal{E} + \frac{\mu_0}{2}\mathcal{H}^\dagger\mathcal{H})d^3x = \int \varepsilon_0 \mathbf{E}^\dagger \mathbf{E} d^3k. \tag{16}$$

III. AM PROPERTIES OF NON-PARAXIAL BEAMS

The AM of a propagating beam in the z-direction is commonly considered in the literature [3, 16, 17, 18, 19] to be equivalent to the line density, that is to say, to the AM per unit length in the z-direction. In order to avoid any possible ambiguity that may arise from the AM density [30, 31], I do not use this notion here. In fact, we have given in Eqs. (14) and (15) the expressions for the spin and orbital AM themselves with respect to the origin. In this section, we will use those expressions to investigate the AM properties of nonparaxial beams. To do this, let us now convert the representation form of nonparaxial beams that was advanced in Ref. [22] into a form that is suitable for present purpose.

A. Description of non-paraxial beams: introduction to a new global unit vector

The electric vector $\mathcal{E}$ of a nonparaxial beam in real space is given by Eq. (3). The electric vector $\mathbf{E}$ in reciprocal space is factorized into three factors [22],

$$\mathbf{E} = m\tilde{\alpha} f, \tag{17}$$

where

$$m = (\ \mathbf{u}\ \ \mathbf{v}\ ) \tag{18}$$

is the mapping matrix, $\tilde{\alpha} = \begin{pmatrix} \alpha_1 \\ \alpha_2 \end{pmatrix}$ is the generalized Jones vector that is assumed to be independent of the wave vector and to satisfy the normalization condition $\tilde{\alpha}^\dagger\tilde{\alpha} = 1$, and f is the electric scalar in reciprocal space. The unit column vectors $\mathbf{u}$ and $\mathbf{v}$ of m represent the two mutually orthogonal states of linear polarization and are defined in terms of the local wave vector $\mathbf{k}$ and a global unit vector $\mathbf{I}$ as follows,

$$\mathbf{u} = \mathbf{v} \times \frac{\mathbf{k}}{k},\ \ \mathbf{v} = \frac{\mathbf{k}\times\mathbf{I}}{|\mathbf{k}\times\mathbf{I}|}, \tag{19}$$

which lead to an important normalization property of the mapping matrix,

$$m^T m = 1, \tag{20}$$

where the superscript T denotes the transpose. Unit vector $\mathbf{I}$ can be specified by its polar angle Θ and azimuthal angle Φ. For the sake of simplicity, let us assume $\mathbf{I}$ to lie in the plane zox, that is to say $\Phi = 0$. In this case, we have

$$\mathbf{I}(\Theta) = \mathbf{e}_x \sin\Theta + \mathbf{e}_z \cos\Theta$$

and the mapping matrix

$$m = \frac{1}{k|\mathbf{k}\times\mathbf{I}|}\begin{pmatrix} (k_y^2+k_z^2)\sin\Theta - k_z k_x \cos\Theta & k k_y \cos\Theta \\ -k_y(k_z\cos\Theta + k_x\sin\Theta) & k(k_z\sin\Theta - k_x\cos\Theta) \\ (k_x^2+k_y^2)\cos\Theta - k_z k_x \sin\Theta & -k k_y \sin\Theta \end{pmatrix}, \tag{21}$$

where $|\mathbf{k}\times\mathbf{I}| = [k^2 - (k_x \sin\Theta + k_z\cos\Theta)^2]^{1/2}$. Due to the symmetry relation $\mathbf{I}(\Theta+\pi) = -\mathbf{I}(\Theta)$, it is postulated throughout this paper that

$$|\Theta| \le \frac{\pi}{2}. \tag{22}$$

A monochromatic beam has a definite wavenumber. It is convenient to use spherical polar coordinates to express the electric scalar as

$$f = \frac{\delta(k-k')}{k^2}\bar{f}(\vartheta,\varphi),$$

where $0 \le \vartheta \le \pi$ and $0 \le \varphi \le 2\pi$. Since $\bar{f}(\vartheta,\varphi)$ is a periodic function of φ with period 2π, a physically allowed function has the following Fourier expansion:

$$\bar{f}(\vartheta,\varphi) = \sum_{l=-\infty}^{\infty} f_l(\vartheta)\exp(il\varphi).$$

In this paper, we consider only one term of the expansion and rewrite the electric scalar as follows,

$$f = \frac{\delta(k-k')}{k^2} f_l(\vartheta)\exp(il\varphi), \tag{23}$$

where the angular-spectrum function $f_l(\vartheta)$ is assumed to be square integrable. In order to use the technique of δ-normalization, the complex conjugate of f is replaced with

$$f^* = \frac{\delta(k-k'')}{k^2} f_l^*(\vartheta)\exp(-il\varphi). \tag{24}$$

For a beam that propagates in the z-direction, its angular-spectrum function satisfies

$$f_l(\vartheta) = 0 \text{ for } \frac{\pi}{2} \le \vartheta \le \pi. \tag{25}$$

Furthermore, if the beam is well-collimated and thus can be paraxially approximated, $|f_l(\vartheta)|$ is sharply peaked at $\vartheta = 0$. The half width $\Delta\vartheta$ of $|f_l(\vartheta)|$ is the divergence angle of the beam.

So obtained $\mathbf{E}$ guarantees that the field vectors $\mathcal{E}$ and $\mathcal{H}$ in Eqs. (3) and (4) satisfy Maxwell's equations. Now that unit real vectors $\mathbf{u}$ and $\mathbf{v}$ are orthogonal to each other, the $\tilde{\alpha}$ that is independent of the wave vector acts as a global characteristic to describe the inner degree of freedom of the beam, the state of polarization. We thus have two independent global quantities, $\mathbf{I}$ and $\tilde{\alpha}$, to describe a beam. It should be pointed out that a physically allowed beam may be a linear superposition of a series of so described beam. They each have their own $\mathbf{I}$ and $\tilde{\alpha}$. The beam that we will consider in this paper is assumed to have definite $\mathbf{I}$ as well as $\tilde{\alpha}$. In the following, we will pay much attention to the effect of these two global characteristics on the orbital AM. Only the AM with respect to the origin will be considered.

B. Orbital AM is dependent on I as well as σ

The longitudinal component of orbital AM with respect to the origin can be turned from Eq. (15) into

$$L_z = \int \frac{\varepsilon_0}{\omega} \mathbf{E}^\dagger (-i\frac{\partial}{\partial\varphi}) \mathbf{E} k^2 \sin\vartheta dk d\vartheta d\varphi \tag{26}$$

in spherical polar coordinates. Hereafter the symbol for the origin will be omitted for the sake of simplicity. By making use of Eq. (17), one has

$$\mathbf{E}^\dagger(-i\frac{\partial \mathbf{E}}{\partial\varphi}) = \tilde{\alpha}^\dagger m^\dagger(-i\frac{\partial m}{\partial\varphi})\tilde{\alpha} f^* f + f^*(-i\frac{\partial f}{\partial\varphi}). \tag{27}$$

When property (20) is taken into account, straightforward calculations yield

$$\begin{aligned} m^\dagger(-i\frac{\partial m}{\partial\varphi}) &= -\hat{\sigma}_3\cos\vartheta + \frac{\hat{\sigma}_3}{2}\frac{\cos\vartheta - \cos\Theta}{1-\cos\Theta\cos\vartheta - \sin\Theta\sin\vartheta\cos\varphi} \\ &\quad + \frac{\hat{\sigma}_3}{2}\frac{\cos\vartheta + \cos\Theta}{1+\cos\Theta\cos\vartheta + \sin\Theta\sin\vartheta\cos\varphi}, \end{aligned} \tag{28}$$

where $\hat{\sigma}_3 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$ is the Pauli matrix. Substituting Eq. (28) into Eq. (27) and noticing Eq. (23), one obtains

$$\begin{aligned} \mathbf{E}^\dagger(-i\frac{\partial \mathbf{E}}{\partial \varphi}) &= (l - \sigma\cos\vartheta) f^* f + \frac{\sigma}{2}\frac{(\cos\vartheta - \cos\Theta) f^* f}{1 - \cos\Theta\cos\vartheta - \sin\Theta\sin\vartheta\cos\varphi} \\ &+ \frac{\sigma}{2}\frac{(\cos\vartheta + \cos\Theta) f^* f}{1 + \cos\Theta\cos\vartheta + \sin\Theta\sin\vartheta\cos\varphi}. \end{aligned}$$

Substituting it into Eq. (26) and considering Eqs. (23) and (24), one finds after performing the integration with respect to variables k and φ

$$\begin{aligned} L_z &= \frac{2\pi\varepsilon_0 l}{k^2\omega}\delta(k-k')\int_0^\pi |f_l(\vartheta)|^2 \sin\vartheta d\vartheta + \frac{2\pi\varepsilon_0\sigma}{k^2\omega}\delta(k-k') \\ &\times \int_0^\pi \left\{ \frac{1}{2}\left(\frac{\cos\vartheta + \cos\Theta}{|\cos\vartheta + \cos\Theta|} + \frac{\cos\vartheta - \cos\Theta}{|\cos\vartheta - \cos\Theta|} \right) - \cos\vartheta \right\} |f_l(\vartheta)|^2 \sin\vartheta d\vartheta. \end{aligned} \tag{29}$$

In obtaining Eq. (29), the following integral formula is used:

$$\int_0^\pi \frac{dx}{1 + a\cos x} = \frac{\pi}{\sqrt{1-a^2}}, \quad (|a|^2 < 1). \tag{30}$$

Substituting Eq. (17) into Eq. (16) and considering Eqs. (23), (24), and (20), one has for the total energy of the beam

$$W = \frac{2\pi\varepsilon_0}{k^2}\delta(k-k')\int_0^\pi |f_l(\vartheta)|^2 \sin\vartheta d\vartheta. \tag{31}$$

It is clear that the orbital AM per unit energy is

$$\frac{L_z}{W} = \frac{l}{\omega} + \frac{\sigma}{\omega}\frac{\int_0^\pi \left\{ \frac{1}{2}\left(\frac{\cos\vartheta+\cos\Theta}{|\cos\vartheta+\cos\Theta|} + \frac{\cos\vartheta-\cos\Theta}{|\cos\vartheta-\cos\Theta|} \right) - \cos\vartheta \right\} |f_l(\vartheta)|^2 \sin\vartheta d\vartheta}{\int_0^\pi |f_l(\vartheta)|^2 \sin\vartheta d\vartheta}. \tag{32}$$

Next let us calculate the transverse component of orbital AM. The x-component is rewritten from Eq. (15) to be

$$L_x = -\int \frac{\varepsilon_0}{\omega}\mathbf{E}^\dagger[k_y(i\frac{\partial}{\partial k_z}) - k_z(i\frac{\partial}{\partial k_y})]\mathbf{E} k^2 \sin\vartheta dk d\vartheta d\varphi. \tag{33}$$

According to Eq. (17), one has

$$\begin{aligned} \mathbf{E}^\dagger[k_y(i\frac{\partial}{\partial k_z}) - k_z(i\frac{\partial}{\partial k_y})]\mathbf{E} &= \tilde{\alpha}^\dagger[k_y m^T(i\frac{\partial m}{\partial k_z}) - k_z m^T(i\frac{\partial m}{\partial k_y})]\tilde{\alpha} f^* f \\ &+ f^*[k_y(i\frac{\partial}{\partial k_z}) - k_z(i\frac{\partial}{\partial k_y})]f. \end{aligned} \tag{34}$$

When property (20) is taken into account, straightforward calculations yield

$$k_y m^T(i\frac{\partial m}{\partial k_z}) - k_z m^T(i\frac{\partial m}{\partial k_y}) = \hat{\sigma}_3 \sin\vartheta\cos\varphi + \frac{\hat{\sigma}_3}{2}\frac{(\cos\vartheta - \cos\Theta)\cot\Theta}{1-\cos\Theta\cos\vartheta - \sin\Theta\sin\vartheta\cos\varphi} + \frac{\hat{\sigma}_3}{2}\frac{(\cos\vartheta + \cos\Theta)\cot\Theta}{1+\cos\Theta\cos\vartheta + \sin\Theta\sin\vartheta\cos\varphi}. \tag{35}$$

Substituting Eqs. (34) and (35) into Eq. (33) and considering the rotation symmetry of f in Eq. (23), one obtains after performing the integration with respect to variables k and φ,

$$L_x = -\frac{\pi\varepsilon_0\sigma}{k^2\omega}\delta(k-k')\cot\Theta\int_0^\pi \left(\frac{\cos\vartheta+\cos\Theta}{|\cos\vartheta+\cos\Theta|} + \frac{\cos\vartheta-\cos\Theta}{|\cos\vartheta-\cos\Theta|}\right)|f_l(\vartheta)|^2\sin\vartheta d\vartheta. \tag{36}$$

In obtaining Eq. (36), formula (30) is used. The x-component of orbital AM per unit energy is thus

$$\frac{L_x}{W} = -\frac{\sigma\cot\Theta}{\omega}\frac{\int_0^\pi \frac{1}{2}\left(\frac{\cos\vartheta+\cos\Theta}{|\cos\vartheta+\cos\Theta|} + \frac{\cos\vartheta-\cos\Theta}{|\cos\vartheta-\cos\Theta|}\right)|f_l(\vartheta)|^2\sin\vartheta d\vartheta}{\int_0^\pi |f_l(\vartheta)|^2\sin\vartheta d\vartheta}. \tag{37}$$

Similar calculations give for the y-component of orbital AM per unit energy

$$\frac{L_y}{W} = 0. \tag{38}$$

Eqs. (32), (37), and (38) are valid for any physically allowed angular-spectrum function $f_l(\vartheta)$. Remembering that the unit vector $\mathbf{I}$ lies in the plane zox, they show that as a vector quantity, the orbital AM with respect to the origin is located in the plane formed by $\mathbf{I}$ and the propagation direction for the rotation-symmetry electric scalar (23). Apart from an l-dependent term in the longitudinal component, the orbital AM is closely dependent on the polarization σ through the unit vector $\mathbf{I}$.

For a beam propagating in the z-direction, property (25) is satisfied. Considering our postulation (22), Eqs. (32) and (37) bocome

$$\frac{L_z}{W} = \frac{l}{\omega} + \frac{\sigma}{\omega}\frac{\int_0^{\pi/2}\left\{\frac{1}{2}\left(1+\frac{\cos\vartheta-\cos\Theta}{|\cos\vartheta-\cos\Theta|}\right) - \cos\vartheta\right\}|f_l(\vartheta)|^2\sin\vartheta d\vartheta}{\int_0^{\pi/2}|f_l(\vartheta)|^2\sin\vartheta d\vartheta}, \tag{39}$$

$$\frac{L_x}{W} = -\frac{\sigma\cot\Theta}{\omega}\frac{\int_0^{\pi/2}\frac{1}{2}\left(1+\frac{\cos\vartheta-\cos\Theta}{|\cos\vartheta-\cos\Theta|}\right)|f_l(\vartheta)|^2\sin\vartheta d\vartheta}{\int_0^{\pi/2}|f_l(\vartheta)|^2\sin\vartheta d\vartheta}, \tag{40}$$

respectively. Eq. (40) indicates that if $\mathbf{I}$ is neither perpendicular nor parallel to the propagation direction, the transverse component of orbital AM does not vanish. Let us discuss the following three cases.

1. $|\Theta| = \frac{\pi}{2}$

This is the case in which $\mathbf{I}$ is perpendicular to the propagation direction. The beam described in this case is uniformly polarized [22] in the paraxial approximation in the traditional sense [32]. In this case, Eqs. (39) and (40) become

$$\begin{aligned} \frac{L_z}{W} &= \frac{l}{\omega} + \frac{\sigma}{\omega} \frac{\int_0^{\pi/2} (1 - \cos\vartheta) |f_l(\vartheta)|^2 \sin\vartheta d\vartheta}{\int_0^{\pi/2} |f_l(\vartheta)|^2 \sin\vartheta d\vartheta}, \\ \frac{L_x}{W} &= 0, \end{aligned} \tag{41}$$

respectively, indicating that the transverse component vanishes and the longitudinal component depends on the polarization. It should be noted that the vanishing transverse component here is just with respect to the origin. With respect to any reference point that is not on the beam axis (the z-axis), the transverse component is by no means equal to zero as is shown by Eq. (12). Furthermore, by making use of paraxial approximation in which $\cos\vartheta$ in the integrand of the numerator can be approximated by unity, $\cos\vartheta \approx 1$, Eq. (41) reduces to

$$\frac{L_z}{W} = \frac{l}{\omega}. \tag{42}$$

Only under so special conditions, is the longitudinal component of orbital AM approximately independent of the polarization. Eq. (42) is exactly the result that was obtained from the consideration of paraxial Laguerre-Gaussian beams [3].

2. $\Theta = 0$

This is the case in which the unit vector $\mathbf{I}$ is parallel to the propagation direction. The beam described in this case is known as cylindrical vector beam [33, 34]. In this case, Eqs. (39) and (40) become

$$\begin{aligned} \frac{L_z}{W} &= \frac{l}{\omega} - \frac{\sigma}{\omega} \frac{\int_0^{\pi/2} |f_l(\vartheta)|^2 \cos\vartheta \sin\vartheta d\vartheta}{\int_0^{\pi/2} |f_l(\vartheta)|^2 \sin\vartheta d\vartheta}, \\ \frac{L_x}{W} &= 0, \end{aligned} \tag{43}$$

respectively. The transverse component vanishes too. But it is seen from Eq. (43) that even in the paraxial approximation, the longitudinal component is not independent of the

polarization and is given by

$$\frac{L_z}{W} = \frac{l}{\omega} - \frac{\sigma}{\omega}. \tag{44}$$

3. $|\Theta| \gg \Delta\vartheta$

A well-collimated beam has a very narrow divergence angle $\Delta\vartheta$. This situation allows us to consider such a case in which $|\Theta| \gg \Delta\vartheta$ is satisfied. The refracted beam that occurred in the spin Hall effect of light [24] was proven [23] to belong to this category. In this case, we have $\cos\vartheta - \cos\Theta > 0$ in the region in which $|f_l(\vartheta)|$ is appreciable. Eqs. (39) and (40) are thus approximated as

$$\begin{aligned} \frac{L_z}{W} &\approx \frac{l}{\omega} + \frac{\sigma}{\omega}\frac{\int_0^{\pi/2}(1-\cos\vartheta)|f_l(\vartheta)|^2 \sin\vartheta d\vartheta}{\int_0^{\pi/2}|f_l(\vartheta)|^2 \sin\vartheta d\vartheta}, \\ \frac{L_x}{W} &\approx -\frac{\sigma \cot\Theta}{\omega}, \end{aligned} \tag{45}$$

respectively. The longitudinal component is almost equal to that in the case of $|\Theta| = \frac{\pi}{2}$. But the transverse component is not equal to zero. Eq. (45) expresses a simple polarization dependence through the unit vector $\mathbf{I}$.

C. Spin is dependent only on the polarization

Substituting Eq. (17) into Eq. (14) and taking Eqs. (23) and (24) into account, one gets

$$\mathbf{S} = \frac{\varepsilon_0 \sigma}{k^2 \omega}\delta(k-k') \int \frac{\mathbf{k}}{k}|f_l(\vartheta)|^2 \sin\vartheta d\vartheta d\varphi. \tag{46}$$

It shows that the transverse component of spin vanishes. The longitudinal component is given by

$$S_z = \frac{2\pi\varepsilon_0\sigma}{k^2\omega}\delta(k-k') \int_0^\pi |f_l(\vartheta)|^2 \cos\vartheta \sin\vartheta d\vartheta. \tag{47}$$

Clearly, the spin AM does not depend on the unit vector $\mathbf{I}$. From Eqs. (47) and (31), it follows that the longitudinal component of spin per unit energy is

$$\frac{S_z}{W} = \frac{\sigma}{\omega}\frac{\int_0^\pi |f_l(\vartheta)|^2 \cos\vartheta \sin\vartheta d\vartheta}{\int_0^\pi |f_l(\vartheta)|^2 \sin\vartheta d\vartheta}, \tag{48}$$

which is valid for any physically allowed angular-spectrum function $f_l(\vartheta)$. For a paraxial beam, $\cos\vartheta \approx 1$ holds and Eq. (48) reduces to

$$\frac{S_z}{W} \approx \frac{\sigma}{\omega}. \tag{49}$$

This is what was obtained from the consideration of paraxial Laguerre-Gaussian beams [3].

D. Total AM

The total AM is the sum of spin and orbital AM. Since the transverse component of spin vanishes, we discuss here only the property of longitudinal component of the total AM. Combining Eqs. (32) and (48) together, one has

$$\frac{J_z}{W}=\frac{l}{\omega}+\frac{\sigma}{\omega}\frac{\int_0^\pi \frac{1}{2}\left(\frac{\cos\vartheta+\cos\Theta}{|\cos\vartheta+\cos\Theta|}+\frac{\cos\vartheta-\cos\Theta}{|\cos\vartheta-\cos\Theta|}\right)|f_l(\vartheta)|^2\sin\vartheta d\vartheta}{\int_0^\pi |f_l(\vartheta)|^2\sin\vartheta d\vartheta}. \tag{50}$$

It is instructive to note that J_z does consist of two parts. One depends only on an integer l, and the other depends only on σ. But the former is not the orbital AM, and the latter is not the spin AM. Eq. (50) is valid for any physically allowed function $f_l(\vartheta)$. When Eq. (25) is taken into account for a beam propagating in the z-direction, it becomes

$$\frac{J_z}{W}=\frac{l}{\omega}+\frac{\sigma}{\omega}\frac{\int_0^{\pi/2} \frac{1}{2}\left(1+\frac{\cos\vartheta-\cos\Theta}{|\cos\vartheta-\cos\Theta|}\right)|f_l(\vartheta)|^2\sin\vartheta d\vartheta}{\int_0^{\pi/2} |f_l(\vartheta)|^2\sin\vartheta d\vartheta}, \tag{51}$$

which clearly shows the impact of the unit vector $\mathbf{I}$. If $\Theta=0$, Eq. (51) reduces to

$$\frac{J_z}{W}=\frac{l}{\omega}, \tag{52}$$

which is independent of the polarization whether the beam is paraxial or not. If $|\Theta|=\frac{\pi}{2}$ on the other hand, one gets from Eq. (51)

$$\frac{J_z}{W}=\frac{l}{\omega}+\frac{\sigma}{\omega}, \tag{53}$$

which is also valid beyond the paraxial approximation. Though the total AM exhibits so simple dependence on l and σ, the first term $\frac{l}{\omega}$ is not the orbital AM and the second one $\frac{\sigma}{\omega}$ is not the spin AM, unless the paraxial approximation holds. It will be shown in the next section that the polarization dependence of L_z for a nonparaxial beam of perpendicular $\mathbf{I}$ is the basis of conversion from spin to orbital AM by a high numerical aperture.

In summary of this section, I have shown that the orbital AM is closely related to the unit vector $\mathbf{I}$. It is due to the impact of $\mathbf{I}$ that the orbital AM is dependent on the polarization. If $\mathbf{I}$ is parallel to the propagation direction, both the spin and orbital AM have only longitudinal components. They are all polarization dependent whether the beam is paraxial or not. But

the total AM does not depend on the polarization. To the best of my knowledge, this is the first time to give the AM expression of cylindrical vector beams. If $\mathbf{I}$ is perpendicular to the propagation direction, the spin and orbital AM also have only longitudinal components. But in the paraxial approximation, the orbital AM is nearly independent of the polarization and is equal to $\frac{l}{\omega}$, and the spin AM is nearly equal to $\frac{\sigma}{\omega}$. If $\mathbf{I}$ is neither parallel nor perpendicular to the propagation direction, the transverse component of orbital AM is not equal to zero. Comparison with the result of Ref. [3] indicates that the unit vector $\mathbf{I}$ of Laguerre-Gaussian beams is perpendicular to the propagation direction.

IV. APPLICATIONS

In this section, I will apply the results obtained before to discuss two different problems. One is the so-called paradox on the spin of circularly polarized plane wave. It will be shown that such a paradox does not exist at all. The other is the conversion of partial spin of a paraxial beam to the orbital AM of the focused beam through a high numerical aperture. The conversion will be shown to root in the polarization dependence of both spin and orbital AM.

A. There is no paradox on the spin of circularly polarized plane wave

The so-called paradox on the spin of circularly polarized plane wave has been the subject of discussion [26, 35, 36] ever since Beth [37] experimentally demonstrated that a circularly polarized plane wave carries spin AM $\hbar$ and was still investigated recently [30, 38, 39, 40, 41]. It states that because the electric and magnetic vectors of a circularly polarized plane wave are perpendicular to the wave vector, its momentum density must be in the propagation direction. As a result, the AM component in the propagation direction must be zero [42] due to the cross product of the position vector with the momentum density. This is contrary to Beth's observation.

As we have shown in Section II, the spin of an electromagnetic field in free space does not come from the part of momentum density that produces the net momentum. Instead, it originates from the other part of momentum density that does not have contribution to the net momentum. From this point of view, it follows that there is no paradox on the spin

of circularly polarized plane wave. After all, what is produced from the momentum density in the propagation direction is the net momentum. In order to elucidate that the spin does not originate from this momentum density, let us make use of Eq. (14) to calculate the AM of a plane wave.

The electric vector of a plane wave in reciprocal space is given by

$$\mathbf{E} = m\tilde{\alpha} f_0 \delta^3(\mathbf{k}-\mathbf{k}'), \tag{54}$$

where $\mathbf{k}'$ is the wave vector of the plane wave. If Eq. (54) is substituted directly into Eq. (14), an infinity will occur. To deal with the infinity, we make use of the technique of δ-normalization as before by replacing $\mathbf{E}^*$ with

$$\mathbf{E}^* = m\tilde{\alpha}^* f_0^* \delta^3(\mathbf{k}-\mathbf{k}''). \tag{55}$$

Substituting Eqs. (54) and (55) into Eq. (14), one gets

$$\mathbf{S} = \frac{\sigma}{\omega}\varepsilon_0 |f_0|^2 \frac{\mathbf{k}}{k}\delta^3(\mathbf{k}-\mathbf{k}'). \tag{56}$$

Similarly, substituting Eqs. (54) and (55) into Eq. (16), one has for the total energy of the wave

$$W = \varepsilon_0 |f_0|^2 \delta^3(\mathbf{k}-\mathbf{k}'). \tag{57}$$

It follows that the spin per photon in the plane wave is

$$\frac{\mathbf{S}}{W}\hbar\omega = \hbar\sigma\frac{\mathbf{k}}{k}, \tag{58}$$

which is entirely along the direction of wave vector $\mathbf{k}$. For circular polarizations $\sigma = \pm 1$, the spin AM per photon is $\pm\hbar$, which is in perfect agreement with Beth's experimental observation. This indicates that when one talked about the paradox on the plane wave's spin, he/she did not realize the role that the momentum density in Eq. (8) plays in the AM. It is very interesting to note that we arrive at the quantum feature [25] of photon's spin by a purely classical approach, from which one might appreciate the nonlocal property of the photon. Since the spin comes from the momentum density that does not produce any momentum on one hand and is stored in the whole real space over which the plane wave spreads on the other, it might be probable that the concept of photon's spin density in real space is physically meaningless [30].

B. Conversion from spin to orbital AM by a high numerical aperture

The incident beam in the AM conversion experiment [15] is LG_0^1, a Laguerre-Gaussian beam. So its unit vector $\mathbf{I}$ is perpendicular to the propagation direction and its parameter l is equal to one, $l = 1$. Before focusing, the spin and orbital AM per unit energy of the paraxial beam in the propagation direction are approximately $\frac{\sigma}{\omega}$ and $\frac{1}{\omega}$, respectively, as Eqs. (49) and (42) show. After focusing, the spin per unit energy of the non-paraxial beam is obtained from Eq. (48) to be

$$\frac{\sigma}{\omega}\frac{\int_0^{\pi/2}|f_l(\vartheta)|^2\cos\vartheta\sin\vartheta d\vartheta}{\int_0^{\pi/2}|f_l(\vartheta)|^2\sin\vartheta d\vartheta},$$

indicating that only a fraction of the incident spin remains in the focused beam, where $f_l(\vartheta)$ now stands for the angular-spectrum function of the focused beam. If the rest of the incident spin

$$\frac{\sigma}{\omega}\left(1-\frac{\int_0^{\pi/2}|f_l(\vartheta)|^2\cos\vartheta\sin\vartheta d\vartheta}{\int_0^{\pi/2}|f_l(\vartheta)|^2\sin\vartheta d\vartheta}\right)$$

is converted into the orbital AM [43], the orbital AM of the focused beam should be

$$\frac{1}{\omega}+\frac{\sigma}{\omega}\left(1-\frac{\int_0^{\pi/2}|f_l(\vartheta)|^2\cos\vartheta\sin\vartheta d\vartheta}{\int_0^{\pi/2}|f_l(\vartheta)|^2\sin\vartheta d\vartheta}\right).$$

This is just the result predicted by Eq. (41). We thus explain the conversion from the spin to the orbital AM on the basis that the orbital AM can be dependent on the polarization. If $\sigma = -1$, the orbital AM per photon is less than $\hbar$. On the other hand, if $\sigma = 1$, the orbital AM per photon is larger than $\hbar$. The authors of Ref. [15] put forward their own theoretical explanation based on the analysis of the longitudinal component of the focused beam's electric vector. Because the longitudinal component of the electric vector is not able to represent the whole beam, they failed to show how the orbital AM of the focused beam depends on the polarization of the incident paraxial beam.

V. CONCLUSIONS AND REMARKS

In conclusion, I put forward a rigorous approach to the separation of the total AM into the spin and orbital AM. This approach is based on the analysis of the momentum density. It was shown that the momentum density can split into two parts. One part that does

not produce any momentum corresponds to the spin. The other part that produces all the momentum corresponds to the orbital AM. The spin defined this way was applied to show that there is no paradox about the spin of circularly polarized plane wave. Apart from the conclusion that the spin is dependent on the polarization, I further showed that the orbital AM is also dependent on the polarization. The polarization-dependent orbital AM was applied to explain the experiment [15] that converted partial spin of the paraxial beam LG_0^1 into the orbital AM of the focused beam through a high numerical aperture.

The unit vector $\mathbf{I}$ was shown to have evident impact on the orbital AM. In the first place, Eqs. (32), (37), and (38) show that the orbital AM is located in the plane formed by $\mathbf{I}$ and the propagation direction. Secondly, Eqs. (28) and (35) show that the polarization-dependent term of orbital AM is determined by the direction of $\mathbf{I}$. when $\mathbf{I}$ is parallel to the propagation direction, the orbital AM is always dependent on the polarization. When $\mathbf{I}$ is perpendicular to the propagation direction, the orbital AM is almost independent of the polarization in the paraxial approximation. These phenomena may imply that the orbital AM is most connected with the polarization, the inner degree of freedom, when $\mathbf{I}$ is parallel to the propagation direction and is least connected with the inner degree of freedom when $\mathbf{I}$ is perpendicular to the propagation direction. In a word, the impact of $\mathbf{I}$ on the orbital AM may offer further insights into the nature of the AM of light.

The author would like to thank Masud Mansuripur of the University of Arizona and Thomas G. Brown of the University of Rochester for their helpful discussions. This work was supported in part by the National Natural Science Foundation of China (60877055 and 60806041), the Science and Technology Commission of Shanghai Municipal (08JC1409701 and 08QA14030), the Shanghai Educational Development Foundation (2007CG52), and the Shanghai Leading Academic Discipline Project (S30105).

---

[1] E. Santamato, B. Daino, M. Romagnoli, M. Settembre, and Y. R. Shen, Phys. Rev. Lett. 57, 2423 (1986).

[2] G. Abbate, P. Maddalena, L. Marrucci, L. Saetta, and E. Santamato, Phys. Scr. T39, 389 (1991).

[3] L. Allen, M. W. Beijersbergen, R. J. C. Spreeuw, and J. P. Woerdman, Phys. Rev. A 45, 8185

(1992).

[4] S. Franke-Arnold, L. Allen, and M. Padgett, Laser & Photon. Rev. 2, 299 (2008).

[5] J. Leach et. al., Phys. Rev. Lett. 88, 257901 (2002).

[6] G. C. G. Berkhout and M. W. Beijersbergen, Phys. Rev. Lett. 101, 100801 (2008).

[7] N. B. Simpson, K. Dholakia, L. Allen, and M. J. Padgett, Opt. Lett. 22, 52 (1997).

[8] M. E. J. Friese, T. A. Nieminen, N. R. Heckenberg, and H. Rubinsztein-Dunlop, Nature (London) 394, 348 (1998).

[9] H. He, M. E. J. Friese, N. R. Heckenberg, and H. Rubinsztein-Dunlop, Phys. Rev. Lett. 75, 826 (1995).

[10] L. Paterson, M. P. MacDonald, J. Arlt, W. Sibbett, P. E. Bryant, and K. Dholakia, Science 292, 912 (2001).

[11] B. Piccirillo, C. Toscano, F. Vetrano, and E. Santamato, Phys. Rev. Lett. 86, 2285 (2001).

[12] B. Piccirillo, A. Vella, and E. Santamato, J. Opt. B: Quantum Semiclass. Opt. 4, S20 (2002).

[13] A. T. O'Neil, I. MacVicar, L. Allen, and M. J. Padgett, Phys. Rev. Lett. 88, 053601 (2002).

[14] V. Garcés-Chávez et al., Phys. Rev. Lett. 91, 093602 (2003).

[15] Y. Zhao et. al., Phys. Rev. Lett. 99, 073901 (2007).

[16] S. J. van Enk and G. Nienhuis, Opt. Commun. 94, 147 (1992).

[17] M. V. Berry, Proc. SPIE 3487, 6 (1998).

[18] S. M. Barnett and L. Allen, Opt. Commun. 110, 670 (1994).

[19] L. Allen, M. J. Padgett, and M. Babiker, Progress in Optics 39, 291 (1999).

[20] S. M. Barnett, J. Opt. B: Quantum Semiclass. Opt. 4, S7 (2002).

[21] C.-F. Li, Opt. Commun. (in review).

[22] C.-F. Li, Phys. Rev. A 78, 063831 (2008).

[23] C.-F. Li, Phys. Rev. A 79, 053819 (2009).

[24] O. Hosten and P. Kwiat, Science 319 (2008) 787.

[25] L. Mandel and E. Wolf, Optical coherence and quantum optics (Cambridge University Press, New York, 1995), Chap. 10.

[26] J. Humblet, Physica 10, 585 (1943).

[27] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, Photons and Atoms (Wiley, New York, 1989), Chap. I.

[28] S. J. van Enk and G. Nienhuis, Europhys. Lett. 25, 497 (1994).

[29] Here $\mathbf{a}^\dagger \mathbf{b}$ denotes the inner product of two complex vectors, $\mathbf{a}$ and $\mathbf{b}$, in the form of matrix multiplication.

[30] L. Allen and M. J. Padgett, Opt. Commun. 184, 67 (2000).

[31] Paula B. Monteiro, Paulo A. Maia Neto, and H. Moyses Nussenzveig, Phys. Rev. A 79, 033830 (2009).

[32] D. N. Pattanayak and G. P. Agrawal, Phys. Rev. A 22, 1159 (1980).

[33] K. S. Youngworth and T. G. Brown, Opt. Express 7, 77 (2000).

[34] C.-F. Li, Opt. Lett. 32, 3543 (2007).

[35] J. M. Jauch and F. Rohrlich, The Theory of Photons and Electrons (Addison-Wesley, New York, 1955).

[36] J. W. Simmons and M. J. Guttmann, States, Waves and Photons (Addison-Wesley, MA, 1970).

[37] R. Beth, Phys. Rev. 50, 115 (1936).

[38] R. I. Khrapko, Am. J. Phys. 69, 405 (2001).

[39] L. Allen and M. J. Padgett, Am. J. Phys. 70, 567 (2002).

[40] A. Stewart, Eur. J. Phys. 26, 635 (2005).

[41] M. Mansuripur, Opt. Express 13, 5315 (2005).

[42] D. Lenstra and L. Mandel, Phys. Rev. A 26, 3428 (1982).

[43] T. A. Nieminen, N. R. Heckenberg, and H. Rubinsztein-Dunlop, arXiv: phys/0408080.

APPENDIX: DERIVATION OF EQS. (14) AND (15)

Let us first derive Eq. (15). Substituting Eq. (7) into Eq. (13), one has

$$\mathbf{L}(0) = \mathbf{L}_1 + \mathbf{L}_2 + c.c., \tag{A.1}$$

where

$$\mathbf{L}_1 = \frac{\varepsilon_0}{4(2\pi)^3} \int d^3k' d^3k \int d^3x \frac{\mathbf{E}' \cdot \mathbf{E}}{\omega} \mathbf{x} \times \mathbf{k} e^{i(\mathbf{k}'+\mathbf{k})\cdot\mathbf{x}} e^{-i(\omega'+\omega)t}, \tag{A.2}$$

and

$$\mathbf{L}_2 = \frac{\varepsilon_0}{4(2\pi)^3} \int d^3k' d^3k \int d^3x \frac{\mathbf{E}' \cdot \mathbf{E}^*}{\omega} \mathbf{x} \times \mathbf{k} e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{x}} e^{-i(\omega'-\omega)t}. \tag{A.3}$$

Upon integrating Eq. (A.2) over the real space and noticing the following properties of Dirac's δ function and its first-order derivative,

$$\delta(t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp(i\omega t)d\omega,\ \ \delta'(t) = \frac{i}{2\pi}\int_{-\infty}^{\infty} \omega\exp(i\omega t)d\omega, \tag{A.4}$$

one obtains

$$\begin{aligned}
\mathbf{L}_1 \ =\ & \frac{\varepsilon_0}{4i}\int (k_y\mathbf{e}_z - k_z\mathbf{e}_y)\frac{\mathbf{E}'\cdot\mathbf{E}}{\omega}e^{-i(\omega'+\omega)t}\delta'(k'_x+k_x)\delta(k'_y+k_y)\delta(k'_z+k_z)d^3k'd^3k \\
+\ & \frac{\varepsilon_0}{4i}\int (k_z\mathbf{e}_x - k_x\mathbf{e}_z)\frac{\mathbf{E}'\cdot\mathbf{E}}{\omega}e^{-i(\omega'+\omega)t}\delta(k'_x+k_x)\delta'(k'_y+k_y)\delta(k'_z+k_z)d^3k'd^3k \\
+\ & \frac{\varepsilon_0}{4i}\int (k_x\mathbf{e}_y - k_y\mathbf{e}_x)\frac{\mathbf{E}'\cdot\mathbf{E}}{\omega}e^{-i(\omega'+\omega)t}\delta(k'_x+k_x)\delta(k'_y+k_y)\delta'(k'_z+k_z)d^3k'd^3k.
\end{aligned}$$

It is changed by eliminating the δ functions into

$$\begin{aligned}
\mathbf{L}_1 \ =\ & \frac{\varepsilon_0}{4i}\int (k_y\mathbf{e}_z - k_z\mathbf{e}_y)\frac{\mathbf{E}(k'_x,-k_y,-k_z)\cdot\mathbf{E}}{\omega}e^{-i[(\omega(k'_x,-k_y,-k_z)+\omega]t}\delta'(k'_x+k_x)dk'_xd^3k \\
+\ & \frac{\varepsilon_0}{4i}\int (k_z\mathbf{e}_x - k_x\mathbf{e}_z)\frac{\mathbf{E}(-k_x,k'_y,-k_z)\cdot\mathbf{E}}{\omega}e^{-i[\omega(-k_x,k'_y,-k_z)+\omega]t}\delta'(k'_y+k_y)dk'_yd^3k \\
+\ & \frac{\varepsilon_0}{4i}\int (k_x\mathbf{e}_y - k_y\mathbf{e}_x)\frac{\mathbf{E}(-k_x,-k_y,k'_z)\cdot\mathbf{E}}{\omega}e^{-i[\omega(-k_x,-k_y,k'_z)+\omega]t}\delta'(k'_z+k_z)dk'_zd^3k.
\end{aligned}$$

Noticing the following property of the derivative of the δ function,

$$\int_{t_1}^{t_2} f(t)\delta'(t-t_0)dt = -f'(t_0),\ \ t_1 < t_0 < t_2, \tag{A.5}$$

and taking transformation (5) into account, the above equation is reduced to

$$\begin{aligned}
\mathbf{L}_1 \ =\ & \frac{\varepsilon_0}{4i}\int \frac{k_y\mathbf{e}_z - k_z\mathbf{e}_y}{\omega}\left(\mathbf{E}\cdot\frac{\partial\mathbf{E}^*}{\partial k_x} + i\frac{k_xt}{\varepsilon_0\mu_0\omega}\mathbf{E}^*\cdot\mathbf{E}\right)d^3k \\
+\ & \frac{\varepsilon_0}{4i}\int \frac{k_z\mathbf{e}_x - k_x\mathbf{e}_z}{\omega}\left(\mathbf{E}\cdot\frac{\partial\mathbf{E}^*}{\partial k_y} + i\frac{k_yt}{\varepsilon_0\mu_0\omega}\mathbf{E}^*\cdot\mathbf{E}\right)d^3k \\
+\ & \frac{\varepsilon_0}{4i}\int \frac{k_x\mathbf{e}_y - k_y\mathbf{e}_x}{\omega}\left(\mathbf{E}\cdot\frac{\partial\mathbf{E}^*}{\partial k_z} + i\frac{k_zt}{\varepsilon_0\mu_0\omega}\mathbf{E}^*\cdot\mathbf{E}\right)d^3k \\
=\ & \frac{i\varepsilon_0}{4}\int \frac{1}{\omega}\mathbf{E}^T(\mathbf{k}\times\nabla_{\mathbf{k}})\mathbf{E}^*d^3k.
\end{aligned}$$

By making the variable replacement $\mathbf{k}\to -\mathbf{k}$, it is changed into a familiar form,

$$\mathbf{L}_1 = \frac{1}{4}\int \frac{\varepsilon_0}{i\omega}\mathbf{E}^\dagger(\mathbf{k}\times\nabla_{\mathbf{k}})\mathbf{E}d^3k. \tag{A.6}$$

Since operator $-i\nabla_{\mathbf{k}}$ is Hermitian, the $\mathbf{L}_1$ in Eq. (A.6) is real. A similar calculation produces from Eq. (A.3)

$$\mathbf{L}_2 = \mathbf{L}_1. \tag{A.7}$$

It is clear that substituting Eqs. (A.6) and (A.7) into Eq. (A.1) will yield Eq. (15).

Then we derive Eq. (14). Substituting Eq. (8) into Eq. (11), one has

$$\mathbf{S} = \mathbf{S}_1 + \mathbf{S}_2 + c.c., \tag{A.8}$$

where

$$\mathbf{S}_1 = -\frac{\varepsilon_0}{4(2\pi)^3}\int d^3k'd^3k\int d^3x\frac{\mathbf{E}'\cdot\mathbf{k}}{\omega}\mathbf{x}\times\mathbf{E}e^{i(\mathbf{k}'+\mathbf{k})\cdot\mathbf{x}}e^{-i(\omega'+\omega)t}, \tag{A.9}$$

and

$$\mathbf{S}_2 = -\frac{\varepsilon_0}{4(2\pi)^3}\int d^3k'd^3k\int d^3x\frac{\mathbf{E}'\cdot\mathbf{k}}{\omega}\mathbf{x}\times\mathbf{E}^*e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{x}}e^{-i(\omega'-\omega)t}. \tag{A.10}$$

Upon integrating Eq. (A.9) over the real space and noticing Eq. (A.4), one obtains

$$\begin{aligned}
\mathbf{S}_1 \;=\; & \frac{i\varepsilon_0}{4}\int(E_y\mathbf{e}_z - E_z\mathbf{e}_y)\frac{\mathbf{E}'\cdot\mathbf{k}}{\omega}e^{-i(\omega'+\omega)t}\delta'(k'_x+k_x)\delta(k'_y+k_y)\delta(k'_z+k_z)d^3k'd^3k \\
+\; & \frac{i\varepsilon_0}{4}\int(E_z\mathbf{e}_x - E_x\mathbf{e}_z)\frac{\mathbf{E}'\cdot\mathbf{k}}{\omega}e^{-i(\omega'+\omega)t}\delta(k'_x+k_x)\delta'(k'_y+k_y)\delta(k'_z+k_z)d^3k'd^3k \\
+\; & \frac{i\varepsilon_0}{4}\int(E_x\mathbf{e}_y - E_y\mathbf{e}_x)\frac{\mathbf{E}'\cdot\mathbf{k}}{\omega}e^{-i(\omega'+\omega)t}\delta(k'_x+k_x)\delta(k'_y+k_y)\delta'(k'_z+k_z)d^3k'd^3k.
\end{aligned}$$

It is changed into, by eliminating the δ functions and taking Eqs. (A.5) and (5) into account,

$$\begin{aligned}
\mathbf{S}_1 \;=\; & \frac{i\varepsilon_0}{4}\int\frac{E_y\mathbf{e}_z - E_z\mathbf{e}_y}{\omega}\mathbf{k}\cdot\frac{\partial\mathbf{E}^*}{\partial k_x}d^3k + \frac{i\varepsilon_0}{4}\int\frac{E_z\mathbf{e}_x - E_x\mathbf{e}_z}{\omega}\mathbf{k}\cdot\frac{\partial\mathbf{E}^*}{\partial k_y}d^3k \\
+\; & \frac{i\varepsilon_0}{4}\int\frac{E_x\mathbf{e}_y - E_y\mathbf{e}_x}{\omega}\mathbf{k}\cdot\frac{\partial\mathbf{E}^*}{\partial k_z}d^3k.
\end{aligned}$$

From the transversality condition $\mathbf{k}\cdot\mathbf{E}^* = 0$, we know that

$$\mathbf{k}\cdot\frac{\partial\mathbf{E}^*}{\partial k_x} = -E_x^*,\;\; \mathbf{k}\cdot\frac{\partial\mathbf{E}^*}{\partial k_y} = -E_y^*,\;\; \mathbf{k}\cdot\frac{\partial\mathbf{E}^*}{\partial k_z} = -E_z^*.$$

$\mathbf{S}_1$ then reduces to

$$\mathbf{S}_1 = \frac{1}{4}\int\frac{\varepsilon_0}{i\omega}\mathbf{E}^*\times\mathbf{E}d^3k, \tag{A.11}$$

which is clearly real. Similarly, $\mathbf{S}_2$ in Eq. (A.10) is found to be real and is equal to $\mathbf{S}_1$,

$$\mathbf{S}_2 = \mathbf{S}_1. \tag{A.12}$$

Substituting Eqs. (A.11) and (A.12) into Eq. (A.8) will yield Eq. (14).